Atmospheric Environment 178 (2018) 148–157

Contents lists available at ScienceDirect

Atmospheric Environment

journal homepage: www.elsevier.com/locate/atmosenv

Trends of atmospheric black carbon concentration over the United Kingdom

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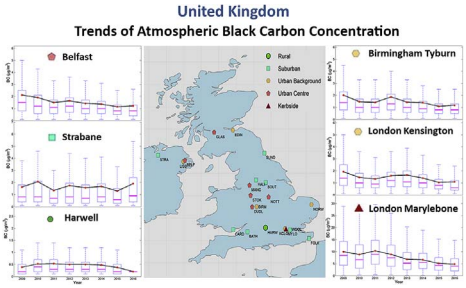
GRAPHICAL ABSTRACT



ARTICLE INFO

Keywords:
Atmospheric black carbon (BC)
BC trends
Diurnal pattern
Emissions
Temporal variations
Climate change

ABSTRACT

The continuous observations over a period of 7 years (2009–2016) available at 7 locations show declining trend of atmospheric BC in the UK. Among all the locations, the highest decrease of 8 ± 3 percent per year was observed at the Marylebone road in London. The detailed analysis performed at 21 locations during 2009–2011 shows that average annual mean atmospheric BC concentration were 0.45 ± 0.10, 1.47 ± 0.58, 1.34 ± 0.31, 1.83 ± 0.46 and 9.72 ± 0.78 $\mu g m^{-3}$ at rural, suburban, urban background, urban centre and kerbside sites respectively. Around 1 $\mu g m^{-3}$ of atmospheric BC could be attributed to urban emission, whereas traffic contributed up to 8 $\mu g m^{-3}$ of atmospheric BC near busy roads. Seasonal pattern was also observed at all locations except rural and kerbside location, with maximum concentrations (1.2–4 $\mu g m^{-3}$) in winter. Further, minimum concentrations (0.3–1.2 $\mu g m^{-3}$) were observed in summer and similar concentrations in spring and fall. At suburban and urban background locations, similar diurnal pattern were observed with atmospheric BC concentration peaks (≈1.8 $\mu g m^{-3}$) in the morning (around 9 a.m.) and evening (7–9 p.m.) rush hours, whereas minimum concentrations were during late night hours (peak at 5 a.m.) and the afternoon hours (peak at 2 p.m.). The urban centre values show a similar morning pattern (peak at 9 a.m.; concentration - 2.5 $\mu g m^{-3}$) in relation to background locations but only a slight decrease in concentration in the afternoon which remained above 2 $\mu g m^{-3}$ till midnight. It is concluded that the higher flow of traffic at urban centre locations results in higher atmospheric BC concentrations throughout the day. Comparison of weekday and weekend daily averaged atmospheric BC showed maximum concentrations on Friday, having minimum levels on Sunday. This study will help to refine the atmospheric BC emission inventories and provide data for air pollution and climate change models evaluation, which are used to formulate air pollution mitigation policies.

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https://doi.org/10.1016/j.atmosenv.2018.01.030
Received 21 April 2017; Received in revised form 13 January 2018; Accepted 16 January 2018
Available online 31 January 2018

1. Introduction

Aerosols are being increasingly recognized as the crucial factor in affecting air quality, climate change and human health. Atmospheric Black Carbon (BC) is a major component of carbonaceous aerosol and is emitted from incomplete combustion of carbonaceous material (Karanasiou et al., 2014). According to World Health Organization report (WHO, 2015), atmospheric BC and other short lived pollutants contribute not only to global warming but also to over 7 million premature deaths related to air pollution (WHO, 2015). Atmospheric BC absorbs solar radiation and thus contributes to warming of the atmosphere as highlighted by Unites States Environmental Protection Agency (USEPA 2012) and (Ramanathan and Carmichael, 2008).

In addition to the global effect, aerosols are associated with severe health effects related to cardiopulmonary and respiratory diseases (Stoeger et al., 2006), and lack of visibility (Jiang et al., 2005). Khan et al. (2006) found that due to its sub-micron size range, atmospheric BC particles has a life time ranging from several days to weeks and can be transported to thousands of kilometers contributing to regional climate change. The long range transport of fine particles can lead to further enhancement of BC concentrations in urban areas which are already influenced by local urban emissions. In a modelling analysis conducted by Singh et al. (2014), it was estimated that the urban increment (including the contributions from urban traffic and other urban sources) was evaluated to be on the average 18%, 33%, 39%, and 43% of the total $PM_{2.5}$ in suburban environments, in the urban background, near roads, and near busy roads, respectively.

The anthropogenic sources of atmospheric BC include vehicular emissions especially from diesel engines, domestic heating, biomass burning and industrial emissions, whereas naturally it is emitted from volcanic emissions and forest fires. The contribution of atmospheric BC emission from fossil fuel, biofuel and open biomass burning which includes forest fires and agricultural waste burning, has been estimated to be ≈ 38%, 20% and 42%, respectively (Bond et al., 2004). It is present in the atmosphere as primary particles and thus can be used as a tracer for combustion sources (Kendall et al., 2001).

Long term measurement of atmospheric BC can be used to estimate its role in regional and global climate change, and also to analyze the temporal variation. Source measurements can be utilized for assessing anthropogenic role in varying atmospheric BC concentration. Several short and long term measurements have been conducted in the past to investigate the seasonal and diurnal pattern of atmospheric BC, in Europe and worldwide (Sahu et al., 2011; Backman et al., 2012; Feng et al., 2014; Tiwari et al., 2013). However, limited number of studies using long term atmospheric BC data have been conducted in this area, in the United Kingdom (UK). Järvi et al. (2008) reported temporal variation of atmospheric BC in Helsinki, Finland in three campaigns with different time scales during 1996–2005 and reported that median atmospheric BC concentration decreased from 1.11 to 1.00 μgm^{-3} during 1996–2005. Saha and Despiau (2009) in their study over Mediterranean coast, Southeast of France during 2005–2006, found that the diurnal variations were more amplified in winter season.

Similarly, Bencs et al. (2010) monitored various pollutants including BC at six sampling sites across Belgium during 2001–2002 for different seasons. Petroleumkaai, one of the site of this study is a famous harbor of Antwerp which is surrounded by petroleum industries. The BC concentration during autumn and winter varies between 0.014–7.4 and 0.025–1.7 μgm^{-3}, respectively. Atmospheric BC were also monitored at the Bight Bank and British Channel shipping route between 2010 and 11 to compare exhaust atmospheric BC concentration with a coastal background site i.e. DeHaan, Belgium. Average BC concentrations at shipping route was 0.366 μgm^{-3} as compared to the DeHaan background site, where BC levels were 0.2 μgm^{-3} (Bencs et al., 2017). Study also report that presence of several oil refining and petroleum industries led to higher values of elemental carbon (EC) at Petroleumkaai than at southern Bight of the North Sea.

Hence, to better estimate the overall atmospheric BC emission load over UK, current study critically assesses the data of real time atmospheric BC observations made during 2009–2016 using UK Black Carbon Network across the UK. This analysis includes 21 stations having various locations i.e. rural, suburban, urban background, urban centre and kerbside. This study specifically focus on seasonal and diurnal variations in atmospheric BC concentrations at all 21 stations during 2009–2011 for better geographical representation and try to understand the atmospheric BC trends over 7 locations during 2009–2016. Variations in atmospheric BC concentrations at different locations were also compared using site specific characteristics and emissions. Further, difference in atmospheric BC concentration during weekday and weekend concentrations were also analyzed. A comparison of the atmospheric BC annual average concentration was also made with other studies including other large scale atmospheric BC measurement networks in Europe, United States, China, Canada, India, Thailand.

Thus the study will be useful in formulating mitigation actions and policy making to curb the atmospheric BC and particulate matter levels. Further, the study also provides data for evaluating global and regional climate models to estimate regional aerosol radiative forcing.

2. Methodology

Atmospheric BC concentrations were measured at 21 locations (Fig. 1) using AE22 Aethalometers make of Magee Scientific (Please see detail in Annexure T1). The network provided ratified AE-22 data based on Virkkula et al. (2007) correction scheme. The BC monitoring sites are part of the UK Black Carbon Network, which is managed and operated for Department for Environment, Food and Rural Affairs

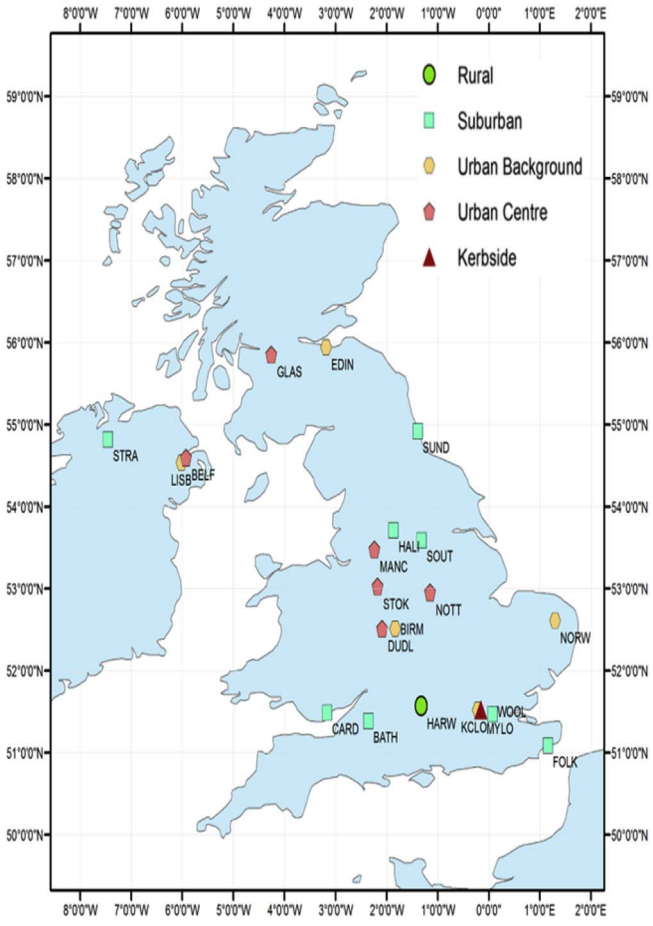

Fig. 1. Map showing atmospheric BC monitoring sites in UK.

Table 1
Location and site specific characteristics of black carbon monitoring sites. (* indicates the stations for which trend analysis has been performed for 2009–2016 data).

SN	Code	Station name	Lon (°N)	Lat (°W)	Type	Country
1	HARW	Harwell*	51.57	−1.33	Rural	England
2	BATH	Bath	51.39	−2.35	Suburban	England
3	CARD	Cardiff*	51.49	−3.16	Suburban	Wales
4	FOLK	Folkestone	51.09	1.16	Suburban	England
5	HALI	Halifax	53.71	−1.86	Suburban	England
6	SOUT	South Kirkby	53.59	−1.31	Suburban	England
7	STRA	Strabane*	54.82	−7.45	Suburban	Northern Ireland
8	SUND	Sunderland	54.92	−1.39	Suburban	England
9	WOOL	Woolwich	51.47	0.07	Suburban	England
10	BIRM	Birmingham Tyburn*	52.51	−1.83	Urban Background	England
11	EDIN	Edinburgh St Leonards	55.95	−3.18	Urban Background	Scotland
12	LISB	Lisburn Dunmurry School	54.54	−6.01	Urban Background	Northern Ireland
13	LOKC	London N. Kensington*	51.52	−0.21	Urban Background	England
14	NORW	Norwich Lakenfields	52.61	1.30	Urban Background	England
15	BELF	Belfast Centre*	54.60	−5.93	Urban Centre	Northern Ireland
16	DUDL	Dudley Centre	52.51	−2.09	Urban Centre	England
17	GLAS	Glasgow Centre	55.86	−4.26	Urban Centre	Scotland
18	MANC	Manchester Piccadilly	53.48	−2.24	Urban Centre	England
19	NOTT	Nottingham Centre	52.95	−1.15	Urban Centre	England
20	STOK	Stoke-on-Trent Centre	53.03	−2.18	Urban Centre	England
21	LOMY	London Marylebone Road*	51.52	−0.15	Kerbside	England

(DEFRA, 2018) (https://uk-air.defra.gov.uk/networks/network-info?view=ukbsn) and by the National Physical Laboratory (NPL). Aethalometers work by collecting aerosol sample every hour on quartz filter tape and measuring the change in attenuation of light at specific wavelength measureing atmospheric BC using optical absorption of light through particulates at specific wavelengths. The data has been analyzed at 21 stations for the period of 2009–2011 (hourly data) having one rural, eight suburban, five urban background, six urban centre and one kerbside station as listed in Table 1. Out of these sites, the measured data at 7 stations have been analyzed for the period of 2009–2016. The data was statistically analyzed using IBM SPSS Statistics 24 package and MATLAB.

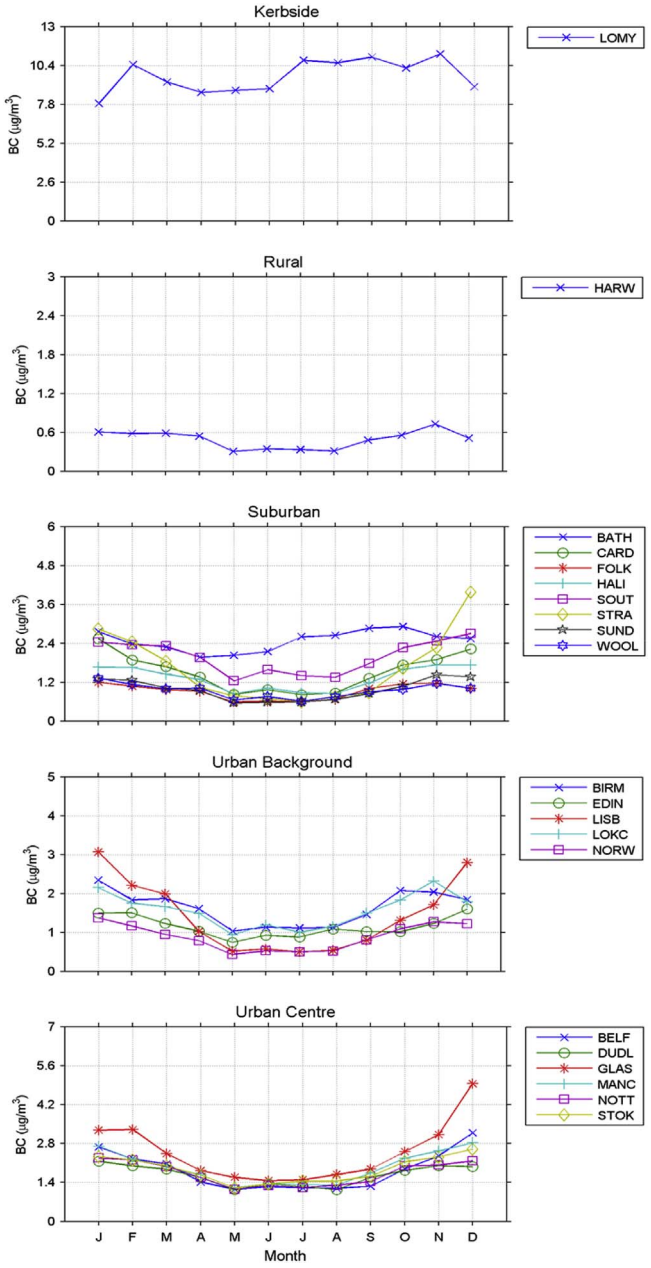

Fig. 2. Mean monthly variation in atmospheric BC concentration at different locations in UK.

3. Results and discussion

3.1. Seasonal variation in atmospheric BC concentration

Seasonal variation in atmospheric BC concentrations for all 21 stations during 2009–2011 has been shown in Fig. 2. At rural location, maximum concentration (0.73 μgm^{-3}) was observed in November, whereas minimum in May (0.30 μgm^{-3}). Monthly mean concentration was found to be 0.49 ± 0.13 μgm^{-3}. At suburban locations, maximum atmospheric BC concentrations were observed during winter season including early spring and varied from 1.2 μgm^{-3} (Sunderland) to ≈ 4 μgm^{-3} (Strabane). Lower atmospheric BC concentrations were observed during summer season ranging from 0.6 μgm^{-3} (Sunderland and Woolwich) to 1.58 μgm^{-3} (South Kirby) as depicted in Fig. 2. Further, at Sunderland lower atmospheric BC concentration was observed (range = 0.6–1.2 μgm^{-3}) throughout the year as compared to other locations. At Strabane, substantially higher peak was observed in December (≈4 μgm^{-3}), due to buildup of pollutant concentration because of stagnant lower wind speed and inversion conditions during extremely low temperatures.

Out of five urban background locations, highest seasonal variations in atmospheric BC concentration were observed at Lisburn, Birmingham and London N. Keningston (Fig. 2), having maximum and minimum concentration in the range of 2.3–3.0 μgm^{-3} and 0.5–1.0 μgm^{-3}, respectively. On the other hand, moderate seasonal variations were observed at Edinburg and Norwich, where atmospheric BC concentration ranges from 0.74 to 1.59 μgm^{-3} and 0.42–1.36 μgm^{-3}, respectively. At urban centre locations, similar seasonal variations were observed for all locations except at Glasgow centre, where extremely high concentration was observed in December (4.95 μgm^{-3}) as compared to other locations having maximum atmospheric BC concentration in the range of 2.2–3.2 μgm^{-3} during late winter (December and January). As mentioned earlier, this is due to extreme low temperatures during winter periods, which leads to stagnant atmospheric conditions causing accumulation of emissions from local sources.

Urban centre during summer season (May to July) have relatively high atmospheric BC concentrations (ranging from 1.2 to 1.5 μgm^{-3}) as compared to suburban/urban background, which is due to the effect of higher traffic emissions at these locations (Fig. 2). At kerbside location (London Marylebone Road) extremely high atmospheric BC concentrations were observed having annual average in the range of 8.8–10.3 μgm^{-3} during 2009–2011. Here, concentrations remained high throughout the year except in the late winters, which is due to less traffic movement during the extreme cold conditions. The distinct pattern of atmospheric BC variation at kerbside shows that the seasonal effect at these sites is overshadowed by traffic congestion.

Seasonal trend of atmospheric BC concentrations shows maximum atmospheric BC concentrations during winter seasons (December and January), which is attributed to stable atmospheric conditions because of lower planetary boundary layer (PBL) resulting in less vertical mixing and accumulation of pollutants close to the earth's surface, lower wind speed and increase in fuel consumption due to space heating, whereas minimum concentrations were observed in summer season, which is attributed to high wind speed and higher PBL resulting in increased vertical mixing or dispersion of pollutants. The large variations of atmospheric BC concentrations are due to data extended over a period of twelve months reflecting various seasons. Ravindra et al. (2008), Ravindra (2011) and Lawrence et al. (2013) also found similar trend for $PM_{2.5}$ and Polycyclic Aromatic Hydrocarbons having 5–7 times higher levels during winter than summer and linked it with incomplete combustion of organic material. The Air Quality Expert Group (2010) also reported similar seasonal variation for $PM_{2.5}$ in the UK.

3.2. Diurnal pattern of atmospheric BC concentration

As shown in Fig. 3, strong diurnal variation in atmospheric BC concentrations were observed at all locations except rural and kerbside, having maximum concentrations during morning (8–9 a.m.) and evening hours (6–9 p.m.) due to high traffic movement during these hours. Supplementary Figure S1 shows mean diurnal variation with standard deviation at different locations of UK. Minimum atmospheric BC concentrations were observed during afternoon (2 p.m.) and early morning hours (5 a.m.). Minimum atmospheric BC concentration was observed during afternoon hour and found to be linked with high wind speeds (shown in Supplementary Figure S2) as observed at suburban, urban background and urban centre locations. Similar morning peak profiles and nocturnal minimum was observed for $PM_{2.5}$ in the UK atmosphere in 2009 (Harrison et al., 2012).

At kerbside, atmospheric BC concentration reached 11 μgm^{-3} at 8 a.m. and reaches its peak value (12.5 μgm^{-3}) around 5 p.m., this indicates the high intensity of traffic congestion throughout the day. The minimum atmospheric BC concentrations i.e. in the range of 5–8 $\mu g\, m^{-3}$ were observed during late night hours (midnight to 5 a.m.), which seems to be linked with limited traffic movement during these hours. At rural location, mild diurnal variation was observed with atmospheric BC concentration varying from minimum 0.35 μgm^{-3} at 6 a.m. to maximum 0.57 μgm^{-3} at 9 p.m. The late night concentrations tend to be higher as compared to the day time due to the accumulation of regional emissions as the PBL decreases. Rural and kerbside locations showed no decrease in afternoon concentration; however the reasons are different for both of these stations. It can be concluded that at rural sites, as there is no influence of morning and evening peaks traffic emissions, the atmospheric BC concentration trend remains almost consistent throughout the day. However, kerbside station seems to be highly influenced by traffic emissions throughout out the day as the site shows no decrease in atmospheric BC concentration during the afternoon (Supplementary Figure S1). Jones and Harrison (2005) also made similar observation, while studying organic and elemental carbon concentrations at rural, urban and kerbside locations of UK. Lower atmospheric BC concentration at rural location also indicate that biomass burning remain restrictive around the site.

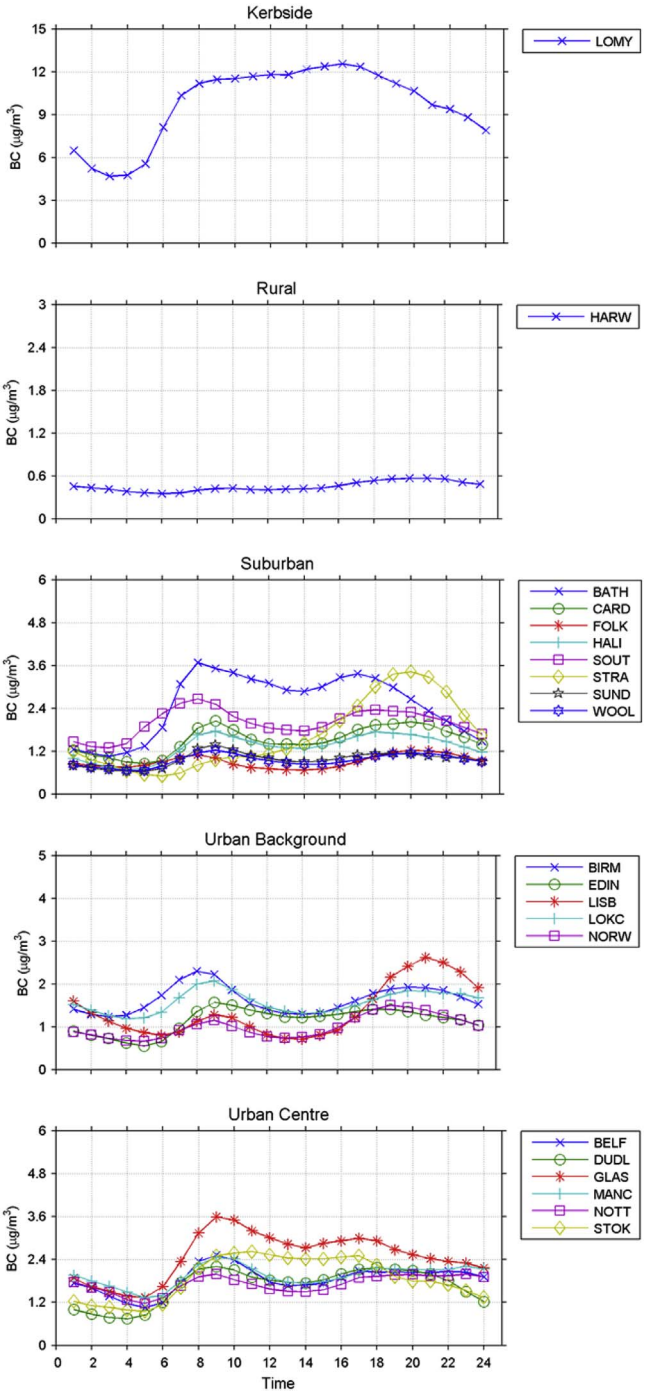

Fig. 3. Diurnal variation in atmospheric BC concentration at different locations in UK.

Similar diurnal pattern was observed at suburban and urban background locations with morning and evening peaks observed at around 9 a.m. and 7–9 p.m., respectively, and minimum concentration during afternoon and early morning hours (Fig. 3). Morning peak seems to be linked with office going commuters and domestic cooking emissions whereas evening peak seems to be influenced by domestic heating and cooking emissions. Further, stable atmospheric conditions during the night may also enhance the local emissions. The Air Quality Expert Group report (2010) also analyzed the diurnal patterns of atmospheric BC concentrations at four locations in UK and suggested that domestic heating can contribute to higher atmospheric BC concentrations apart from morning and evening rush hours traffic emissions. At most of the suburban locations, similar diurnal patterns were observed i.e. atmospheric BC concentration reaching maximum in the morning (between

8 and 9 a.m.) and in evening (between 5 and 8 p.m.), and minimum during the afternoon (around 2 p.m.) and late night hours (3–5 a.m.).

Although, the range of maximum concentrations observed varied largely from 1.2 μgm^{-3} at Folkstone and Sunderland to 3.68 μgm^{-3} at Bath. A distinct pattern was observed at Strabane, where no peak in concentration was observed in the morning hours. However, a constant increase in atmospheric BC concentration was observed from 6 a.m. (0.5 μgm^{-3}) to 8 p.m. (3.4 μgm^{-3}) and seems to be linked with the dominance of domestic heating emissions (Fig. 3). South Kirby and Bath showed higher atmospheric BC concentrations as compared to other locations with a maximum of 2.5 μgm^{-3} and 3.6 μgm^{-3}, respectively, which was found comparable with maximum atmospheric BC concentration observed at urban centre locations. This shows that atmospheric BC emission levels in Bath are similar to that at urban centre locations. South Kirby also had higher concentrations during morning hours between 5 and 10 a.m. and then in the evening hours between 4 and 8 p.m.

Urban background locations showed similar diurnal patterns at suburban locations with maximum atmospheric BC concentrations observed during morning (between 8 and 9 a.m.) and evening hours (between 7 and 9 p.m.), whereas minimum concentrations during afternoon hours (between 1 and 2 p.m.). Diurnal variation and concentration level of atmospheric BC at Birmingham and London were almost same with relatively higher atmospheric BC concentrations as compared to other locations as shown in Fig. 3, which is due to higher traffic emissions at these locations. A distinct peak was observed during evening at Lisburn (2.6 μgm^{-3}) as compared to morning (1.3 μgm^{-3}), which suggests high domestic heating emissions.

Among urban centre locations, Dudley centre, Belfast, Manchester Picadily and Nottingham Centre showed similar atmospheric BC concentration and diurnal variation. Morning peaks were observed at 9 a.m. (2–2.5 μgm^{-3}), whereas concentration remained at around 2 μgm^{-3} from 5 p.m. to 11 p.m. This is linked with traffic movement as the traffic count remains high at these locations till late night hours. Highest atmospheric BC concentrations were observed at Glasgow during morning (3.6 μgm^{-3}) and evening peaks (3 μgm^{-3}) with afternoon minima (2.7 μgm^{-3}) greater than the maximum concentration observed at other urban centre locations. Stoke-on-Trent showed a continuous high atmospheric BC concentration (around 2.5 μgm^{-3}) from 9 a.m. to 5 p.m. which shows constant high traffic conditions during the time period. Minimum concentrations were observed during afternoon at 2 p.m. and late night hours between 4 and 5 p.m.

3.3. Comparison of weekday vs weekend atmospheric BC concentrations

Daily mean atmospheric BC concentrations for each day of a week were averaged for the period 2009–2011, and the trend is shown in Fig. 4 for each category of location. Concentrations on weekdays were found to be significantly higher as compared to weekend, which can be attributed to high vehicular emissions on weekdays. Highest variation was observed at kerbside location with atmospheric BC concentration declining from 11.2 μgm^{-3} on Friday to 6.64 μgm^{-3} on Sunday. This could be predicted as vehicular emissions were identified as predominant source, which decreases significantly due to decrease in vehicular movement from weekdays and weekend. Further, other locations are affected by sources such as cooking, residential heating or other site specific sources. On the other hand, lowest variation was observed at rural location with a mild difference of 0.1 μgm^{-3} between weekday and weekend atmospheric BC concentration which could be due to less use of vehicles.

Large variation was observed at urban centre with atmospheric BC concentration declining from over 2 μgm^{-3} on Friday to less than 1.5 μgm^{-3} on Sunday (Fig. 4). Further, urban background and suburban locations shows slightly less variation of 0.38 and 0.55 μgm^{-3}, respectively (Fig. 4). Highest atmospheric BC concentration was observed on Friday, whereas lowest on Sunday. This suggests that as the week progresses, atmospheric BC concentration starts accumulating near the surface level due to the vehicular movement of office going commuters till Friday, and then declines till Sunday.

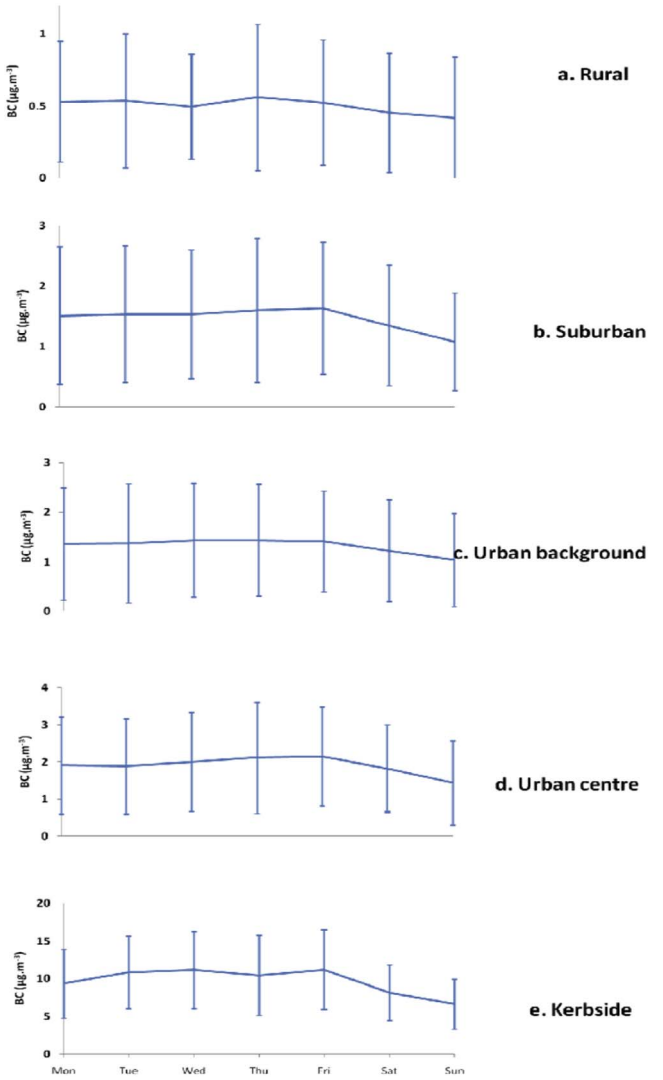

Fig. 4. Average (± standard deviation) atmospheric BC concentrations over weekdays and weekends at different locations in UK, 2009–2011.

3.4. Variation in atmospheric BC concentration at different site types

Annual average, maximum and minimum atmospheric BC levels observed at different locations viz. rural, suburban, urban background, urban centre and kerbside during 2009–2011 are shown in Table 2. Concentrations at the rural location increased from 0.34 $\mu g\, m^{-3}$ in 2009 to 0.51 $\mu g\, m^{-3}$ in 2010 and 2011, whereas a moderate decrease in concentrations were observed at suburban (0.2 μgm^{-3}), urban background (0.45 μgm^{-3}) and urban centre (0.67 μgm^{-3}) from 2009 to 2011. Annual average atmospheric BC concentration at kerbside decreased by over 1 μgm^{-3} from 2009 to 2010 but increased in 2011 by 1.5 μgm^{-3} to reach 10.34 μgm^{-3}. Similar concentrations were observed among suburban and urban background locations in 2009 (1.57 μgm^{-3}) and 2010 ($\approx$1.4 μgm^{-3}), whereas in 2011 urban centre concentration declined to suburban levels i.e.$\approx$ 1.4 μgm^{-3}. Overall, highest concentrations were observed at kerbside followed by urban centre, suburban, urban background and rural locations as shown in Table 2. Further analysis of 3 year data (2009–2011) is depicted in Fig. 5 for various sites. From Fig. 5, it could be interpreted that around

Table 2
Annual average atmospheric BC concentration according to location subclass in UK during 2009–2011.

Station	Descriptive statistics of BC concentration (μg.m^{-3})									Mean (2009–2011)
	2009			2010			2011			
	Avg ± SD	Min	Max	Avg ± SD	Min	Max	Avg ± SD	Min	Max	
Harwell	0.34 ± 0.26	0.03	1.14	0.51 ± 0.38	0.01	2.33	0.51 ± 0.50	0.04	3.01	0.45
Bath	2.51 ± 1.16	0.40	8.19	2.20 ± 1.11	0.32	6.11	2.74 ± 1.19	0.36	6.13	2.48
Cardiff	1.61 ± 1.23	0.29	9.36	1.53 ± 1.07	0.27	7.35	1.38 ± 1.04	0.20	8.40	1.51
Folkestone	0.96 ± 0.69	0.12	5.31	0.81 ± 0.58	0.08	3.39	0.96 ± 0.87	0.10	7.31	0.91
Halifax	1.54 ± 1.11	0.23	7.39	1.33 ± 1.12	0.23	11.31	1.07 ± 0.81	0.20	5.04	1.31
South kirkby	2.22 ± 1.51	0.26	12.86	2.00 ± 1.64	0.19	14.37	1.76 ± 1.29	0.17	9.25	1.99
Strabane	1.57 ± 1.19	0.16	7.23	2.03 ± 2.45	0.09	18.86	1.29 ± 1.24	0.23	10.68	1.63
Sunderland	1.06 ± 0.92	0.08	5.87	0.91 ± 0.75	0.01	5.00	0.92 ± 0.73	0.14	4.41	0.96
Woolwich	1.07 ± 0.70	0.16	5.28	0.94 ± 0.56	0.18	3.69	0.81 ± 0.51	0.13	3.20	0.94
Birmingham Tyburn	2.01 ± 1.46	0.25	9.00	1.47 ± 1.03	0.16	7.28	1.42 ± 1.07	0.20	9.30	1.64
Edinburgh St Leonards	1.31 ± 0.86	0.21	6.84	1.15 ± 0.69	0.15	4.62	0.97 ± 0.58	0.10	4.18	1.14
LisburnDunmurry High School	1.34 ± 1.36	0.12	7.23	1.70 ± 2.09	0.14	13.11	1.11 ± 1.50	0.13	11.00	1.38
London N. Kensington	1.93 ± 1.20	0.41	8.47	1.45 ± 1.15	0.19	11.26	1.33 ± 1.00	0.26	9.48	1.57
Norwich Lakenfields	1.28 ± 0.87	0.20	3.75	0.99 ± 0.77	0.13	6.37	0.74 ± 0.53	0.09	3.11	1.00
Belfast Centre	2.12 ± 1.46	0.42	9.50	1.90 ± 1.78	0.29	14.63	1.50 ± 1.22	0.17	10.15	1.84
Dudley Centre	1.66 ± 1.03	0.21	5.50	1.9 ± 1.09	0.43	8.09	1.35 ± 0.87	0.11	6.58	1.65
Glasgow Centre	2.94 ± 1.85	0.63	14.49	2.75 ± 2.29	0.32	18.79	1.77 ± 1.20	0.18	9.79	2.49
Manchester Piccadilly	2.07 ± 1.22	0.48	10.84	2.05 ± 1.42	0.53	10.77	1.71 ± 1.14	0.26	10.72	1.94
Nottingham Centre	1.89 ± 1.09	0.33	7.30	1.78 ± 1.06	0.32	8.17	1.48 ± 0.98	0.20	5.90	1.71
Stoke-on-Trent Centre	2.02 ± 1.17	0.39	8.40	2.07 ± 1.20	0.41	8.78	1.50 ± 0.95	0.14	8.66	1.87
London Marylebone Road	9.99 ± 4.71	1.34	22.75	8.84 ± 5.19	1.18	22.62	10.344.61	1.35	21.96	9.72
Mean ± SD of 21 stations	2.07 ± 1.91			1.92 ± 1.68			1.75 ± 2.02			

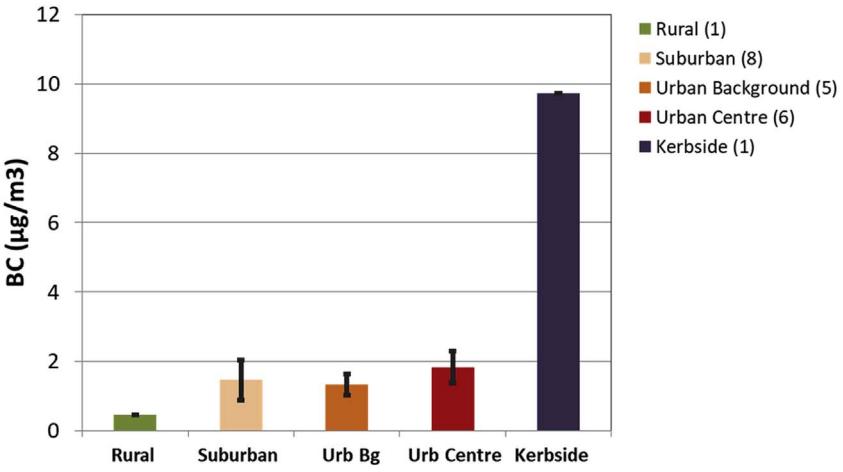

Fig. 5. Observed average (2009–2011) BC concentration at Rural, Suburban, Urban Background, Urban Centre and Kerbside stations. The vertical bars show standard deviation. The numbers in the bracket show the number of stations in each type.

Table 3
Trend values of atmospheric BC concentration and percentage decline every year.

S. No.	Station Name	Type	Trend (μgm^{-3})	Trend (%)	p value (alpha = 0.05)
	Belfast Centre	Urban Centre	−0.13 ± 0.05	−6.13 ± 2.4	0.0006
	Birmingham Tyburn	Urban Background	−0.10 ± 0.08	−4.95 ± 4.0	0.026
	Cardiff	Suburban	−0.11 ± 0.02	−6.83 ± 1.2	0.0002
	Harwell	Rural	−0.03 ± 0.04	−7.69 ± 10.3	0.1386
	London Marylebone Road	Kerbside	−0.80 ± 0.32	−8.02 ± 3.2	0.0008
	London N. Kensington	Urban Background	−0.09 ± 0.08	−4.66 ± 4.1	0.0275
	Strabane	Suburban	−0.01 ± 0.11	−0.62 ± 6.8	0.7944

1 μgm^{-3} can be attributed to urban emissions, whereas 8 μg m^{-3} can be attributed to traffic in Marylebone road near the roads.

3.5. Trend analysis of atmospheric BC concentration over UK

Annual average atmospheric BC concentration data was available for a period of 2009–2016 for 7 stations (viz. urban background, urban centre, suburban, rural, kerbside) and it was analyzed to study the trend of annual average atmospheric BC concentrations over UK. The application of f-test statistic shows statistically significant decrease in atmospheric BC concentrations except at rural (Harwell) and suburban (Strabane) (Table 3). As shown in Fig. 6, the concentration trend shows fall over a period of 2009–2016 except a moderate increase in the year 2011 at rural (Harwell) and kerbside (London Marylebone) and again

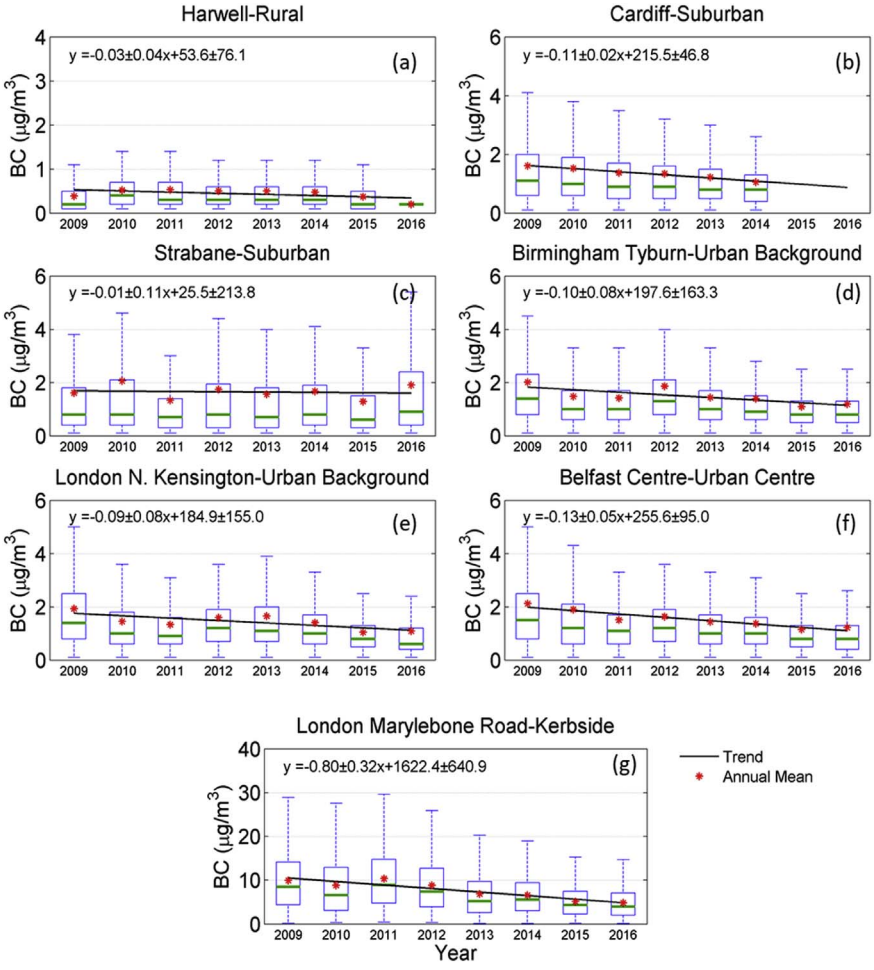

Fig. 6. Box-Whisker plot of hourly atmospheric BC concentration from 2009 to 2016 at seven locations in the UK. The green line shown the median value, the box shows the inter quartile range. The red star shows the annual mean and the black line shows the trend. The equation shows the linear regression parameters. Extremely high values (outliers) have not been shown here. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

slight decrease at suburban site in 2016 compared to 2009. Overall, among the 7 stations data the highest decline (8.03 ± 3.2 percent per year) in BC concentration was observed at the kerbside site i.e. London Marylebone whereas, marginal decline (0.62 ± 6.8 percent per year) in BC levels were recorded at the suburban site i.e. Strabane. According to the report Cleaner Air for London (2013), black carbon monitored at three sites showed statistically insignificant small decrease in BC concentrations. However, current study shows statistically significant decline in BC concentration over UK, which could be linked with various policy interventions taken at local and national level in the UK. These policy measures include - promotion of sustainable mode of transport (walking and cycling initiatives), introduction of Euro IV standards for buses, zero tailpipe emission capable taxis and hybrid diesel-electric buses among others. On the other hand, development of Ultra Low Emission Zone (ULEZ), new emission standards for construction equipment, updating the Clean Air Act were also announced to improve air quality of London (GLA, 2013). Font and Fuller (2016) also examined BC at 65 locations in London and observed 11% per year decrease in BC concentrations between 2010 and 2014. In agreement with the current study, the London study also links decline in BC concentrations with the various policy initiatives taken by Greater London Authority.

3.6. Atmospheric BC concentration with meteorological variables

Variation in BC concentration against wind for different type of station has been shown in Fig. 7. The meteorological data for 2010 was taken as indicative meteorological analysis, which was obtained at nearby station from Met Office Integrated Data Archive System (MIDAS) British Atmospheric Data Centre (BADC) data. The corresponding meteorological station has been shown in Table 4. As shown in Fig. 7 (and in Supplementary Figure S3, S4), it could be observed that wind plays important role in controlling the atmospheric BC concentration at all type of stations except that Marylebone road which is a kerbside station. Marylebone Road is one of the busiest roads of central London with several street canyons. This site is highly influenced by local traffic emissions and because of the street geometry; wind speed shows almost no influence on the BC concentrations. Jones et al. (2010) also studied the dependence of wind speed on pollutant concentrations at kerbside, rural and urban locations and observed that general relationship between wind speed and pollutant concentration did not hold true for kerbside as the complexity of street canyon and the buoyancy effects of exhaust emissions restricted free flow of wind.

3.7. Comparative overview of atmospheric BC concentration

Large scale network for measurement of atmospheric BC concentration has been established in several developed and developing countries such as United States, European Union, Canada and China. Table 5 shows the range of annual mean atmospheric BC concentration observed at urban and rural locations in various large scale atmospheric BC measurement networks and single location studies. As in the present study, significant difference in atmospheric BC has been found between urban and rural locations. In the present study, the annual mean concentrations observed during 2009–2011 at 19 urban and rural locations in United Kingdom were in the range (0.9–2.5) μgm^{-3} and 0.45 μgm^{-3},

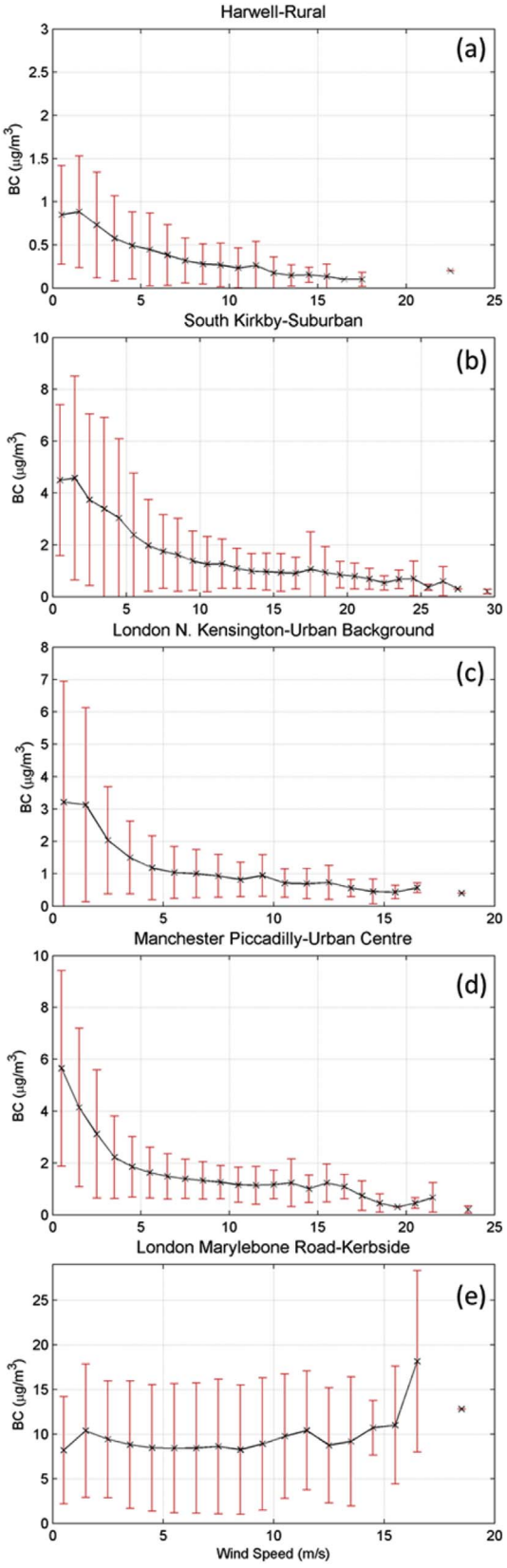

Fig. 7. Graphical representation of atmospheric Black carbon concentration with wind speed at different locations in UK for data collected in 2009.

respectively, which is comparable to the atmospheric BC concentrations observed in Europe (EMEP, EUSAAR), US (CSN and IMPROVE) and Canada (NAPS), France (Saha and Despiau, 2009) and Finland (Järvi et al., 2008) despite of different methods of atmospheric BC measurement such as thermal and optical. However, much higher atmospheric BC concentrations were observed in various studies conducted at several locations in China (CAWNET, Feng et al., 2014; Cao et al., 2009; World Health Organization, 2015) and India (Kant et al., 2012; Guha et al., 2015; Rastogi and Sarin, 2009; Ramachandran and Kedia, 2010; Mahapatra et al., 2014). Relatively lower concentrations were found in Europe (1.4–1.8 $\mu g\, m^{-3}$) for annual average of atmospheric BC monitored during 2002-03 since 11 of the 14 background sites taken for EMEP campaign were regional background sites. Comparatively higher concentrations were observed in winter season due to residential heating as compared to summer season (Yttri et al., 2007; USEPA 2012).

Table 4
Met station corresponding to a BC station.

BC Stations	Met Stations	Distance (km)
Manchester Piccadilly	MANCHESTER HULME LIBRARY	1.8
London N. Kensington	KEW (ROYAL BOTANIC GARDENS)	6.8
London Marylebone Road	KEW (ROYAL BOTANIC GARDENS)	10.4
SOUTH KIRKBY 1	BRAMHAM	30.6
Harwell	WISLEY	65.7

4. Conclusions

This is the first detailed study of long term trends of atmospheric BC concentration several locations in the UK during 2009–2016 and the application of f-test shows statistically significant decrease in atmospheric BC concentrations except at Harwell (rural) and Strabane (suburban) locations. The highest decline of 8 ± 3 percent per year is found at the kerbside site Marylebone road in London followed by rural site Harwell and suburban site Cardiff having a decline of 7.69 ± 10.3 and 6.83 ± 1.2 percent per year respectively. The suburban Strabane and urban background site London N. Kensington show marginal decreasing trend of 0.62 ± 6.8 and 4.66 ± 4.1 percent per year respectively. The diurnal and seasonal analysis has been performed at 21 locations during 2009–2011. Highest monthly average atmospheric BC concentration was observed during winter season (e.g. December, 2.52 μgm^{-3}) and were found to be almost twice that of summer levels (e.g. May, 1.31 μgm^{-3}). High concentrations during winter season was attributed to stable atmospheric conditions, lower PBL and increase in fuel consumption due to space heating. Lower concentrations in summer were found to be associated with high wind speed and higher PBL resulting in increased vertical mixing or dispersion of pollutants. Diurnal variation showed high atmospheric BC concentrations during the morning (8–9 a.m.) and evening (6–9 p.m.) hours and lower atmospheric BC levels were observed during afternoon (2 p.m.) hours due to higher vehicular emissions by office going commuters during morning and evening.

Site specific analysis shows that maximum atmospheric BC concentration was at kerbside (9.72 ± 0.78 μgm^{-3}) followed by urban centre (1.83 ± 0.46 μgm^{-3}), suburban (1.46 ± 0.58 μgm^{-3}), urban background (1.34 ± 0.30 μgm^{-3}) and having lowest at rural location (0.45 ± 0.10 μgm^{-3}). It is shown that almost 1 μgm^{-3} is attributed to urban emissions most likely because of domestic cooking and biomass combustions, whereas traffic contributed up to 8 $\mu g\, m^{-3}$ of atmospheric BC near busy roads where vehicular movement was predominant source. Highest daily average atmospheric BC concentration was observed on Friday whereas lowest on Sunday at all locations due to decrease in traffic count. Difference between weekday and weekend concentrations were highest at kerbside (5.6 μgm^{-3}) followed by urban centre (0.71 μgm^{-3}), suburban (0.55 μgm^{-3}), urban background (0.38 μgm^{-3}) and the least at rural location (0.11 μgm^{-3}). Interestingly, atmospheric BC concentration remain high at kerbside throughout the year except in the late winters, which is due to less

Table 5
Comparison of atmospheric BC concentrations reported over the globe.

Country (Measurement network/Studies)	Method	Year	Range of annual average atmospheric BC concentration (μgm^{-3})	
			Urban	Rural
United States (CSN, IMPROVE) [a]	Thermal Optical	2005–07 2007	0.3–2.5 (≈200 sites) 0.3–3.0 (≈45 sites)	0.1–0.6 (≈150 sites)
Canada (NAPS) [a]	Thermal	2003–09	0.9–1.8 (12 sites)	0.4–0.8 (4 sites)
Europe (EMEP) [a] (Yttri et al., 2007)	Thermal	2002–03	1.4–1.8 (2 sites)	0.2–1.8 (12 sites)
Europe (EUSAAR)[a]	Thermal Optical	2006	1.5 (2 sites) 2.7 (1 site)	0.1–0.7 (4 sites) 0.2–0.5 (4 sites)
Goonhilly, Cornwall, (UK Butterfield and Quincey, 2017)	Optical	2012–2013	–	0.22 (1 site)
Europe (6 cities) (Reche et al., 2011)	Optical and Thermal	2009	1.7–1.9 (background) (2 sites) 3.5–7.8 (traffic) (2 sites) 0.7–0.8 (shipping, industrial)	–
Milan, Italy (Europe) (Invernizzi et al., 2011)	Optical	July (year of data collected not mentioned)	1.5–2 (pedestrian-only zone) 2.6–3.1 (intermediate traffic) 3.3–6.3 (high traffic)	–
Barcelona, Spain (Europe) (Pérez et al., 2010)	Optical	2007	3.6 ± 1.8 (0.2–25.3)	–
LIFE + MED-PARTICLES Project (Barcelona & Athens) (Ostro et al., 2015)	ThermalOptical	2008–09; 2009–10	0.739 (Athens; 2008–09) 2.011 (Barcelona; 2009–10)	–
China (CAWNET) [a]	Thermal	2006	9.3–14.2 (5 sites)	0.3–5.3 (13 sites)
Shanghai, China (Feng et al., 2014)	Optical	2010–11	3.3–3.8 (1 site)	–
Dehradun, India (Kant et al., 2012)	Optical	2007	4.3 ± 0.62 (1 site)	–
Agartala, India Guha et al., 2015	Optical	2010–12		Winter- 17.8 ± 9.2 Monsoon- 2.8 ± 1.7
Arabian sea, India Babu et al., 2004 [c]	Optical	2003	March- 0.7 June- 0.1	–
Western India Rastogi and Sarin, 2009	Thermal		0.3–4.4	–
Ahmedabad, India Ramachandran and Kedia, 2010	Optical	2008	1.6–13.8	–
Bhubaneshwar, India Mahapatra et al., 2014	Optical	2010–12	1.56–11.55	–
Bangkok, Thailand (Sahu et al., 2011)	Thermal	2007–08	3.0–4.3 (1 site)	–
China Cao et al., 2003	Thermal	2003	9.9 in winter 3.6 in summer (14 sites)	–
Toulon, France Sahu et al., 2011	Optical	2005–06	0.3–1 (1 site)	–
Helsinki, Finland Järvi et al., 2008 [b]	Optical	1996–97 2000–01 2004–05	1.11 0.93 (1 site) 1.00	–
Shanghai and Beijing, China (Zhou et al., 2009)	Optical	2005 (Summer)	Beijing- 2.37 Shanghai- 5.47	–
United Kingdom (BC network) (Present study)	Optical	2009–11	0.91–2.49 (19 sites)	0.45 (1 site)

[a] USEPA 2012, Report to Congress on Black Carbon.
[b] Concentrations are reported as median concentrations during the study period.
[c] Measurements were conducted using aircraft during March–April 2003 and May to June 2003 over Arabian sea.

traffic in extreme cold conditions and seasonal holidays. Hence, it is inferred that seasonal effect is overshadowed by high vehicular emissions. The annual average atmospheric BC concentrations at all sites except kerbside in UK were in the range of 0.3–2.94 μgm^{-3}, which was found to be comparable with the atmospheric BC concentrations reported in several studies in Europe but shows a declining trend for a period of 2009–2016. These trends are most likely linked to local and national policy initiatives (e.g. promotion of sustainable mode of transport, strict emission standards from transport and non-transport sector, and development of ULEZ)

Acknowledgements

RK would like to thank Department of Health Research, Indian Council of Medical Research, Ministry of Health and Family Welfare via Letter number V-25011/25(19)2013/HR dated 16 October 2014, for providing the Fellowship Training Programme in Environmental Health under Human Resource Development Health Research Scheme.

Appendix A. Supplementary data

Supplementary data related to this article can be found at http://dx.doi.org/10.1016/j.atmosenv.2018.01.030.

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